

A Review of Background Ozone in the Troposphere

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1. Introduction

Ozone (O_3) is found at all levels in the atmosphere. In the stratosphere it plays an extremely important and beneficial role in screening the lower layers of the atmosphere and the surface of the earth from the sun's harmful ultraviolet radiation. In the troposphere, and especially at the surface of the earth, ozone is not beneficial. It has been shown to be harmful to human health, vegetation and certain materials. Ozone in the troposphere is also a significant greenhouse gas. This review concentrates on ozone in the lower atmosphere, sometimes referred to as ground-level ozone. For this reason the location designation will not be included in the following discussion. In specific cases, if necessary to avoid confusion, ozone will be described as troposphere or stratospheric, but without this designation troposphere ozone is to be understood.

Ozone is not emitted directly to the atmosphere, but is formed in reactions between oxides of nitrogen (NO_x , which is the sum of NO , nitric oxide, and NO_2 , nitrogen dioxide) and volatile organic compounds (VOC), these reactions being driven by absorbed solar radiation. Significant anthropogenic emissions of oxides of nitrogen occur in association with combustion processes, e.g., from motor vehicles, thermal power generation and industrial and other boilers. Volatile organic compound emissions occur wherever there is incomplete combustion, e.g., from motor vehicles, and also in the manufacture, processing and use of such compounds, e.g., in the petrochemical industry, motor fuel production and distribution and solvent use.

There are also natural sources of NO_x , such as production by lightning and direct emission from soils, and of VOC, mainly by emission from vegetation. In the context of ozone production carbon monoxide, CO , is included as a VOC. It has a relatively low reactivity to ozone formation, but this is compensated for by its high abundance, relative to other VOC. Methane, CH_4 , also has relatively low reactivity but high abundance. Methane and CO contribute to ozone production in the polluted atmosphere, but are of particular importance in the photochemical production of ozone in the free troposphere.

Measures for the reduction of ozone involve reducing the emissions of both NO_x and VOC to the atmosphere. The situation is complicated by the highly non-linear nature of the atmospheric reactions which lead to the formation of ozone. In other words, a reduction of X% in emissions does not necessarily produce an X% reduction in ozone concentrations. Indeed, it is not at all uncommon, notably in highly urbanized areas, for a reduction in NO_x emissions to lead to a local increase in ozone concentrations. This happens because emissions of NO_x in such areas can react with ozone already present, leading to reduction in concentration, a phenomenon generally referred to as ozone scavenging or titration. However, concentrations of ozone further downwind will increase as a result of the increased NO_x emissions. If the emissions of NO_x are then reduced there will be less reaction, leaving more ozone in the local atmosphere, though the overall effect beyond the local area will probably be a reduction in ozone concentration.

A further complication in the design of ozone control measures is the presence of a significant concentration of background ozone. Precursor emissions in the region of concern lead to production of additional ozone, which is approximately additive to the background. However, it is only the local emissions which are controllable by any jurisdiction, meaning that the background concentration is the lowest level that can be achieved.

Policy discussions in several jurisdictions, including Ontario, refer to controllable ozone. As discussed above, this is the component of the locally occurring ozone which can be controlled by controlling emissions within the jurisdiction.

The US EPA defines Policy Relevant Background ozone (OAQPS, 2007), which is the ozone concentration which would be observed in the absence of emissions of anthropogenic precursors in the US, Canada or Mexico. Specifically, Policy Relevant Background ozone includes ozone from natural precursor emissions in the US, Canada and Mexico, and ozone arising from the transport of ozone or its precursors, emitted from both natural and anthropogenic sources, from outside the US, Canada and Mexico.

2. Background Ozone

a. Sources

In any region measurable, sometimes significant, concentrations of ozone will be found, even in the total absence of local emissions. This background concentration of ozone will generally have both a natural and an anthropogenic component.

The natural component results from the reaction of naturally emitted NO_x and VOC. Nitrogen oxides are naturally emitted from soils, as a consequence of microbial processes occurring in the soil, and may also be produced by lightning and by forest fires (many of which are started by lightning strikes). The predominant source for biogenic (natural) VOC is vegetation. Vegetatively emitted VOC, mainly pinenes and terpenes, are highly reactive in the formation of ozone, and the magnitude of their emissions increases with increased sunlight and temperature, just the conditions which favour ozone production. Additional natural sources of VOC, mainly methane, are emissions from wetlands and geological and geothermal leakage from underground deposits.

It is also possible for ozone to be formed directly in the atmosphere by lightning.

Another possible source for naturally produced ozone in the troposphere is the stratosphere. Although the stratosphere and the troposphere are generally decoupled, with little transfer across the tropopause (the boundary between the two), conditions do arise in which such transfer can occur. This phenomenon may be the result of a fold in the tropopause, leading to the formation of a “bubble” of stratospheric air, rich in ozone, which becomes trapped in the troposphere. Bubbles of this type usually remain well above the surface, but may occasionally be brought to lower levels.

Ozone in layers of the troposphere above the boundary layer can have a lifetime of many days or even a week or two. This is because the major loss processes, scavenging by nitric oxide and dry deposition, occur at or very close to the surface of the earth. This means that ozone produced in one region can, if lifted to higher levels, travel to another region, contributing there to the uncontrollable background for that region. Lifting mechanisms include deep convection in the atmosphere and orographic lifting (i.e., lifting of air parcels as the land surface rises to higher altitudes). It has been shown (Fiore et al., 2002; Jaffe et al., 2003) that western North America receives a background ozone contribution from Asia and Europe, while Auvray and Bey (2005) have demonstrated that Europe receives ozone transported from both North America and Asia.

b. Determination of Background

The most obvious approach to follow in determining background ozone in a region, is to make measurements that are not impacted by emissions from the region itself. This will require that measurements be made in remote locations, usually at or near the region's boundary. In addition, appropriate protocols will be required to ensure that the measurements used to determine the background do not include any influence from the region of interest. Typically, this would involve considering measurements made when the wind is blowing only from the appropriate sector. Greater certainty would be achieved by ensuring that the back trajectories associated with the measurement times lie only within the appropriate sector, thus avoiding cases where the trajectory starts out over the study region, then turns enough that it reaches the measurement site from the clean sector (Carslaw, 2005; Nolle et al., 2001; Simmonds, et al., 2004).

Naja et al. (2003) determined background ozone concentrations at two high elevations locations in Europe, using a time of travel approach, in which each ozone concentration measurement was associated with the residence time of the corresponding air parcel over Europe. Ozone concentration was correlated with residence time, and extrapolated back to zero residence time. The corresponding concentration was interpreted as background, entering the continent.

Lefohn and coworkers have done considerable work on background ozone (e.g., Altshuller and Lefohn, 1996; Lefohn et al. 2001; Oltmans et al., 2006). The methods that they have used to delineate the background concentration include making measurements at background sites, particularly using measurements made at such sites in February, when little photochemical production of ozone is expected. They have, in other analyses, made use of concurrent measurements of photochemically produced chemical species such as PAN (peroxyacetyl nitrate) and reactive organic nitrates to identify when the measured ozone contains a contribution from local and regional photochemistry in addition to pure background. Many of their analyses have included the use of back trajectories to characterise air mass origins.

Simultaneous measurements of other markers for polluted air, such as carbon monoxide, NO_x or halogenated organic compounds have been used for diagnostic purposes (e.g.,

Simmonds et al., 2004; Scheel et al., 1997). Ozone concentrations associated with low concentrations of the marker or markers are designated as background. A variant of this approach is to assume that the NO_x concentrations at a low percentile of the overall concentration distribution are characteristic of background air parcels (Bronnimann, et al., 2000). Values for this threshold have been variously set in the range of 2 to 10%. Munger et al. (2002) used a similar method, but required either CO or NO_x to be in the lower 10% of their concentration distribution.

Fiore et al. (2002, 2003) have used a 3-dimensional global tropospheric model with chemistry (GEOS-CHEM) to investigate background ozone in North America, using two different approaches. GEOS-CHEM incorporates a parameterisation of stratospheric ozone input, using the SYNOZ method (Connell et al., 2001; Logan, 2006). Model verification has included testing of this parameterisation against ozonesonde measurements, and ²¹⁰Pb-⁷Be-O₃ correlations obtained during aircraft flights. The first method followed tagged tracers to separate ozone produced within North America from that produced outside the region. The tracer is a notional chemical species, which is subject to the same chemical and physical loss rates as ozone, but which originates from a specific geographic area. Ozone concentrations within the study domain were then separated, or deconstructed, according to the relative concentrations of tracer originating outside and inside the domain.

In the second approach, emission scenarios were modelled involving setting all anthropogenic emissions to zero globally (Scenario 1), within North America (Scenario 2) and outside North America (Scenario 3). Scenario 1 provides an estimate of the natural contribution to background ozone in North America, while Scenario 2 was used to estimate the contribution of anthropogenic emissions outside North America to the background. Scenario 3 allows estimation of the same contribution in a different way, and was included since comparing the two estimates provides a measure of the impact of non-linearity in the formation and removal of ozone. The impact of non-linearity was found to be small.

The GEOS-CHEM model has similarly been used to study background contributions to ozone in Europe (Auvray and Bey, 2005).

c. Reported Values of Background

Background ozone concentrations found in the course of this literature review are summarised in Table 1. These concentrations were determined at many locations, almost all in the northern hemisphere, covering a wide range of topography and using a variety of methods to isolate background concentrations. Not surprisingly, therefore, a relatively wide range of background concentrations is listed in the table.

Background ozone levels were reviewed by Vingarzan (2004). The reported data were collected at sites characterised as “background”, and ranges of annual median and annual maximum concentrations were reported. Many of the median concentrations were in the approximate range of 20 to 40 ppb, while the maxima ranged up to 116 ppb. Many of the

stations reported maximum ozone concentrations exceeding 100 ppb. These observations strongly suggest that the measurement sites listed in this review are not free of local or regional scale anthropogenic influence, as is confirmed by consideration of the site locations. For example, the Egbert, Ontario site is less than 100 km from Toronto, while Kejimikujik, in Nova Scotia, is impinged at times by polluted air parcels travelling up the east coast from Boston and other heavily urbanised areas further south.

A summary of background ozone concentrations was provided by Fiore et al. (2003). The results of five research groups were quoted, values ranging overall between 25 and 45 ppb.

McKendry (2006) reported background ozone concentrations for British Columbia, building on the strong base provided by the review by Vingarzan (2004). He found that the background concentration was in the range 20 – 35 ppb, with the maximum occurring in the spring. Little evidence was found for a stratospheric contribution, but trans-Pacific transport was found to contribute as much as 5 to 15 ppb.

It may be noted that background concentrations reported for North America are similar to those found in Europe. This is particularly true if concentrations determined at high elevation are treated as a separate group, since ozone concentrations tend to increase with altitude. For example, Jaffe et al. (2003) have reported background concentrations ranging from 32 to 44 ppb for six coastal California sites, while Altshuller and Lefohn (1996) suggested that background ozone concentrations in the US were 35 ± 10 ppb. In comparison, Simmonds et al. (2004) in a careful analysis of measurements made at Mace Head, on the west coast of Ireland, determined that an appropriate range for the background ozone concentration was 39.3 ± 6 ppb.

Other European results have been reported by Lindskog and Kindbom (2001) and Scheel et al. (1997), and have also been reviewed and summarised by ACCENT (2006). (ACCENT is the Atmospheric Composition Change Network of Excellence funded by the European Community). These studies found background ozone concentrations in the range 25 to 40 ppb.

Analysis of measurements made at an elevated site in Japan (Pochanart et al., 2004) indicated that the background ozone concentration was 44.4 ± 3.6 ppb. Of this background, approximately 10 ppb came from east Asia.

Thus, it may be suggested that there is a central tendency to the values given in Table 1 of approximately 25 to 40 ppb for the sites at lower elevation. However, a discrepancy remains between the values reported recently by Lefohn and coworkers (e.g., Cooper et al., 2005; Lefohn et al., 2001), which are significantly higher than those determined using the GEOS-CHEM model (Fiore et al., 2003).

In their recent publications, Lefohn et al. (2001) and Cooper et al. (2005) maintain that significant inputs occur of air of stratospheric origin, bringing with it ozone concentrations of 60 ppb or higher. On the other hand, research done using a modelling

approach with the GEOS-CHEM model supports background ozone concentrations that are much lower, often in the range 25 to 35 ppb.

Such a large discrepancy has significant implications for policy development, since if the background concentration is indeed 60 ppb, extremely stringent controls would be required to achieve a standard of 80 ppb, the current US value. On the other hand, the results of Fiore et al., suggest that the background concentration is actually depressed under conditions which give rise to ozone episodes. In other words, the controllable portion of the ozone is higher than previously believed, and therefore more amenable to reduction. These and other issues will be addressed in Section 2e, Discussion.

d. Trends in background ozone

Measurements of ozone at ground level have been made since the 1850s, at which time it may be assumed that essentially all of the ozone was background. It is difficult to establish the accuracy of the early measurements, particularly because of their susceptibility to interference from other atmospheric constituents. However, a considerable effort was made by Marengo et al. (1994) to form a dataset that was as consistent as possible, for comparison with modern measurements. This work was subsequently extended by Gros (2006), and the resultant temporal pattern is shown in Figure 1.

As is shown in the figure, modern levels of ozone are approximately five times those reported in the mid nineteenth century. Even if the early measurements are in error by a factor of two, this still indicates a significant trend. It must be remembered that the measurements shown in Figure 1 are for high altitude locations, which means that the absolute values are higher than the concentrations that will be found closer to sea level. Nevertheless, the entire data set is consistent with respect to the altitude of the measurement sites; in particular, the earliest (lowest concentration) and the latest (highest concentration) measurements were made at the same location, namely Pic du Midi.

In her review of background ozone, Vingarzan (2004) also reported trends for a number of locations worldwide. Data for twenty two of thirty stations showed increasing trends ranging from near zero to 2.6% per year, with the majority of the annual increases being about 0.2 to 0.5% per year. Those sites at which ozone has decreased, reported rates in the range from close to zero to 1.4% per year. It should be noted that while the sites for which results are reported are characterised as “background”, it is not always clear that the measurements relate strictly to background ozone. For example, the largest rate of decrease (1.4% per year) was reported for Edmonton, Alberta. Given the magnitude of precursor emissions over much of the province of Alberta, it would require very careful analysis to isolate the effect of purely background ozone at this site.

Trend rates for a number of other monitoring locations have been retrieved from the current literature, and are reported in Table 2. Lindskog and Kindboom (2001) found little significant change for ozone concentrations measured at remote locations in northern Scandinavia. However, Naja et al. (2003) report that background ozone

concentrations have increased by 5 to 10 ppb over the 30 year period from 1970 to 1999, corresponding to an annual rate of increase of approximately 0.15 to 0.3 ppb.

Results for high altitudes (above 3000 m in Switzerland) presented by Bronnimann et al. (2000) indicate an overall increase of 0.8 to 1.3 ppb over 9 to 11 years. This corresponds to approximately 0.1 ppb per year.

Analysis of measurements made close to sea level show an increase of 1 ppb over 4 years for Gozo, Malta, though, as discussed previously, these measurements probably include the influence of the industrialised and urbanised areas of the Mediterranean basin. At Mace Head, on the west coast of Ireland, Simmonds et al. (2004) found an annual rate of increase in background ozone of 0.49 ppb per year. However, this value is higher than the value reported by Carslaw (2005) for the same location of 0.18 ppb.

Trends in background ozone for North American locations are found in the work of Jaffe et al. (2003), who reported an annual average increase of 0.5 ppb. The measurements from which this trend was derived were made in California. Lin et al. (2000) found a statistically significant rate of increase of background ozone over 20 years of 3 to 5 ppb for the Spring and Fall. The greatest rate of increase was in the northeastern US.

Although the review by Vingarzan (2004) identified some decreasing trends in background ozone, none of the additional literature found in the present review has reported a decreasing trend in background ozone. Apart from one report of no significant increase, only increasing rates were found. The rate of increase ranged from approximately zero to 0.5 ppb per year. If the background ozone concentration is approximately 35 ppb, this corresponds to a rate of up to 1.4% per year. Many of these trends have been consistent over a number of years, at least ten in many cases, but as many as 30 for the results presented by Naja et al. (2003). These trends were obtained for a number of locations across the northern hemisphere, with isolated measurements also available for the southern hemisphere. There does not appear to be a significant location dependence for the trend rates, implying that these ranges would apply specifically to North America.

The rates of change may well be different in the future, but projections for future years prepared by the International Panel on Climate Change, and reported by Vingarzan (2004), suggest that background ozone concentrations will continue to increase through 2040, 2060 and 2080, but then decreasing slightly to 2100. The low end of the projected range increases from about 34 to 40 ppb, with the high end increasing from 50 ppb in 2040 to about 85 ppb in 2080, then decreasing slightly to 84 ppb in 2100. The corresponding average projected values are about 42 ppb in 2040, 54 ppb in 2060, 63 ppb in 2080 and 62 ppb in 2100 (see Table 4 for a summary).

e. Discussion of Background Ozone Concentrations and Trends

A number of observations may be made concerning the background ozone concentrations and trends reported in the preceding two sections:

- “Background” is an apparently simple concept, but different researchers have different understanding or definition of the term, sometimes making it difficult to compare results from different studies. For example, Nolle et al. (2001) reported background concentrations greater than 50 ppb, as measured on the island of Gozo, Malta. These concentrations represent the background for Malta, since care was taken to analyse only measurements made for wind trajectories which did not include Malta itself. However, Malta is located in the Mediterranean, just south of Sicily. It is close to major land-based emission sources, besides being adjacent to major shipping lanes, so that the concentrations measured here should be considered representative of the anthropogenically influenced atmosphere of the Mediterranean basin
- The assumption is often made that measurements made at high elevation are representative of the free troposphere (the portion of the troposphere above the mixing layer), and may therefore be accepted as representing background. Care must be taken in accepting this assumption, since upward mixing may occur to a greater extent than expected (Bonasoni et al., 2000), because of convective or orographic lifting, which delivers boundary layer air to such locations. In any event, in interpreting results from high elevation sites it must be remembered that ozone concentrations tend to increase with increasing altitude.
- Substantial disagreement exists between background ozone concentrations reported by two groups working in the United States. As may be seen in Table 1, Lefohn and co-workers (Lefohn et al., 2001; Cooper et al., 2005) maintain that background ozone concentrations in North America, and elsewhere can exceed 60 ppb, with substantial contributions to this amount coming from the stratosphere. They have reached this conclusion on the basis of a detailed analysis of measured ozone concentrations, with various methods used to segregate out the measurements which correspond to background conditions. On the other hand, the group at Harvard (Fiore et al., 2003) have applied a global atmospheric model with chemistry to derive background concentrations in the range of 25 to 35 ppb, with occasional excursions to higher values. It must be accepted that both methods are subject to some uncertainty. For example, direct analysis of measurements relies on the exclusion of contributions from regionally polluted air masses, while the modelling approach of Fiore et al. relies on the accuracy of the model and its predictions. However, it appears likely that a major part of the discrepancy is related to the question of what exactly is understood by background. The Lefohn et al. results are necessarily obtained from sites at or near the edge of the study domain, so that a clean sector or sectors can be defined. The GEOS-CHEM model predicts concentrations at all points within the study domain as an integral consequence of the simulations. In particular, Fiore et al. believe that the meteorological conditions which often lead to high ozone concentrations in eastern North America (i.e., large scale stagnation) cause the background ozone entering the domain to be substantially reduced by deposition and local scavenging. Thus for an ozone episode in which the maximum

concentration was 80 ppb or higher, they found a background contribution of only 15 ppb or less for the majority of episodes. The implications of this result for policy design are very important, since it means that a larger proportion of local ozone than often expected is controllable by reducing regional emissions. On the other hand, if the higher value is correct, the background concentrations are often high enough that there is essentially no hope of reducing local emissions to the point that compliance with current ozone standards or guidelines could be achieved.

- A number of reports have indicated that maximum ozone concentrations have reduced over the last several years, responding to reductions in the emissions of the precursors, NO_x and VOC. However, concentrations at the lower end of the frequency distribution have tended to increase. This is interpreted as being reflective partly of a worldwide increase in background concentrations, but is also believed to be a consequence of reduced NO_x emissions, resulting in reduced reduction of ozone concentrations by chemical scavenging (Wallasch, 2003; Chevalier et al., 2007).
- It was noted in much of the literature surveyed that concentrations of ozone produced anthropogenically in the local region tend to peak in the summer, whereas natural background concentrations tend to peak in the spring. The latter observation is believed to be a consequence of a pulse of natural emissions released at the start of the growing season, but may also include a contribution from stratospheric input. It is interesting that Carslaw (2005) suggests that the seasonal profile measured at Mace Head, on the western coast of Ireland, has changed in recent years from one having a summer maximum to a spring maximum, indicating that emission reductions in Europe have decreased anthropogenic production to the point that the major component of ozone measured at the site is background.
- There is wide agreement that ozone can be transported from one continent to another. Thus up to 5 or 10 ppb of ozone produced from North American precursor emissions can reach Europe, with a similar amount from Asia reaching western North America (Fiore et al., 2002; Vingarzan, 2004; McKendry, 2006). This is of great importance, given the rapid industrialisation of China and India, which will almost inevitably lead to increases in emissions from the region. It is clear that international agreement(s), beyond what is under discussion between Canada and the US, will be required for success in the control of ozone.
- The literature reviewed has provided information on long term trends of background ozone, and also on the seasonal cycles of concentration. However, no information was found on shorter term variations in background concentration. For example, Lefohn et al. maintain that significant contributions arise from intrusions of stratospheric air, but it is not clear how long the background concentration will remain elevated following such an event. It is also not clear what sort of diurnal pattern will be followed by background ozone. It is known that there is often very little by way of a diurnal cycle at remote locations, but it is important also to know how the background concentration varies through the day at locations which are also influenced by locally and regionally produced ozone. This knowledge would be required, for example, when separating the health costs

of ozone due to local and regional emissions from those purely due to background.

- With a few exceptions noted above, the current literature indicates that background ozone concentrations are increasing, and will continue to do so until the end of the 21st century. It is very difficult to determine how much of the increase is in the anthropogenic component and how much is purely natural. The modelling work of Fiore et al. (2003) suggests that the natural component is generally in the 10 to 25 ppb range, and never exceeds 40 ppb. Compare this range with the concentrations measured at Pic du Midi in the mid-nineteenth century (Figure 1), which were approximately 10 to 15 ppb. If it is assumed that these concentrations are accurate, and that they contain no anthropogenic contribution, it may be inferred that there has been a small increase in the natural component of the background. Simmonds et al. (2004) suggest that some of the increase in background ozone concentrations may be associated with increasing burning of biomass in the tropical and boreal regions. However, at least some of this burning should be considered to be anthropogenic.

f. The Ontario Context

Background ozone has been specified and used in two different ways in Ontario (Bloxam, 2007). The two applications are discussed below.

First: In modelling the production and transport of photochemical smog in the region, the Models-3/CMAQ system is applied to a domain extending from west of Chicago to the east coast, and from the northern end of Florida as far north as James Bay. Models-3/CMAQ is a current-generation, regional-scale model, originally developed by the US EPA, and now widely used in many jurisdictions including Ontario. The MOE model domain encompasses essentially all of the pollutant sources which have an impact on Ontario, but their impact is superimposed on a background of ozone entering through the boundaries of the model domain. These boundary conditions are specified as an input to the model, including an altitude dependence. The values in the approximately 300 metre deep layer at the surface are set to 35 ppb on the southern and western boundaries, rising to 40, 45, 50, 60 and then 70 ppb through five further levels to the top of the model domain. Slightly lower values are set for the northern and eastern boundaries, e.g., 30 ppb in the lowest layer.

The boundary condition values are based on profiles observed in ozone measurements made over many years by agencies such as Environment Canada. The profiles used in the CMAQ modelling are based on worldwide measurements archive by Environment Canada (the data are archived by the World Ozone and Ultraviolet Radiation Centre, accessible at http://www.woude.org/index_e.html), and are consistent with profiles determined in many other studies (e.g., Pisano et al., 1997; Zeng et al., 2006).

Second: Mathematical modelling of photochemical air pollution is used for many purposes. In the particular application where results are used to determine the deleterious

impacts and associated costs of ozone on human health, crops and materials, a separate determination of background has been used. This value is needed throughout the model domain to allow separation of the impacts due to ozone produced from precursor emissions within the domain, from those associated with background and naturally occurring ozone. The component due to emissions in the domain is variously referred to as Controllable Ozone or Policy Relevant Ozone.

The Ontario background concentrations were determined by carrying out a model run with all anthropogenic emissions within the domain shut off. The ozone concentrations that were produced in this run were the result of the boundary conditions and the biogenic emissions within the domain. Most of the values were in the range of 35 to 40 ppb, with some as low as 30 ppb, and some exceeding 50 ppb. These 8-hour average values varied over space, and also from day to day, as biogenic emissions and meteorological and transport conditions varied. These values are consistent with those reported by Galvez and Sloan (2004)

From this discussion it is apparent that background ozone concentrations observed and used in Ontario are consistent with worldwide experience in two major respects.

- Firstly, the concentrations found are comparable with those found in other locations.
- Secondly, there are two distinct “backgrounds”, namely the boundary conditions to the Ontario modelling domain, and the non-controllable portion of the ozone throughout the domain. This non-controllable portion will contain both biogenic and anthropogenic (from outside the domain) components.

In the light of background ozone concentrations reported in current literature, the boundary condition concentrations used in mathematical modelling in Ontario are appropriate. However, it would be worthwhile to develop an understanding of why the non-controllable concentrations determined within the domain are somewhat higher than those reported by Fiore et al. (2003).

3. Survey of Ozone Standards and Guidelines

Many jurisdictions worldwide have established standards or guidelines for troposphere ozone. These standards represent upper limits to the ambient concentration, which are intended to protect sensitive receptors. Most standards are set to protect human health, but some have been established to be protective of crops.

The science of determining a “safe” level for a pollutant such as ozone is very complex, and is beyond the scope of the current review. Indeed, current science indicates that there is no lower limit below which no damage is inflicted on at least some part of the population. As a result, other considerations also enter into the setting of standards, encompassing the feasibility of achieving the desired level. Included in the assessment of feasibility, though not necessarily stated explicitly, is balancing between the cost of the measures required to achieve the concentration limit, and the costs of the damages which result if this level is exceeded.

Also to be borne in mind in determining the feasibility of achieving a standard is the effect of background. As has been previously discussed, the background concentration in any jurisdiction is a level below which local controls can not reduce the concentration. The background is composed of both natural and anthropogenic components, with the latter resulting from intercontinental transport, meaning that some part of the background is potentially controllable if international agreement can be reached on limiting the production of ozone worldwide. Nevertheless, there remains the natural component, which is not amenable to control.

For the reasons outlined above there tends to be some divergence between the standard or guideline values set in different jurisdictions. These values are summarised for selected jurisdictions in Table 3. In addition to setting a numerical value for the standard, a specification is normally also attached defining how the measurements are to be compared with the standard. Thus in Canada and the US the standard for ozone is specified as an 8-hour running average, with a numerical value of 65 ppb and 0.08 ppm respectively (noting that the US standard (US EPA) is currently undergoing a review and revision process). For each calendar year at a given monitoring site the fourth highest value of this average is taken and an average is formed of the fourth highest values over three successive years. The three year average is then tested against the standard. The Canada-wide standard is to be achieved by 2010. The Canadian and US standards are intended to be protective of human health.

Note that because of rounding, the ozone metric in the US does not exceed the standard until it reaches 85 ppb. These standards are intended to be protective of human health. The US has set a secondary standard, protective of vegetation, which has the same numerical value and specification as the primary standard.

The ozone standard for Mexico is expressed as a daily maximum one-hour concentration, not to be exceeded, of 0.110 ppm. This standard is intended to protect human health.

The European Union (European Union, 2002) in Directive 2002/EC/3 mandated an ozone standard to be achieved by 2010. The metric is an 8-hour concentration of $120 \mu\text{g m}^{-3}$ or approximately 60 ppb. Numerically this is a relatively stringent standard, relative to those of other jurisdictions, though the limit may be exceeded as many as twenty five times per calendar year, with the number of exceedences being averaged over three years. It is intended however that there be no more than one exceedence per year by 2020.

A one-hour ozone standard of 0.06 ppm has been set in Japan. This may be the most stringent standard currently in place worldwide. It has, however, also been necessary to establish a warning level of 0.12 ppm and a serious warning level of 0.24 ppm, with an emergency level of 0.40 ppm (all one-hour values).

Air quality standards for China are set for three area classes:

Class I:	Residential
Class II:	Commercial
Class III:	Industrial

The corresponding numerical values, which are all expressed as one-hour averages, are 0.12, 0.16 and $0.20 \mu\text{g m}^{-3}$, corresponding to approximately 60, 80 and 100 ppb, but noting the possible inclusion of numerical rounding as is the case with the US standard.

Australia is apparently unique in adopting a 4-hour average for its ozone standard. The numerical value is 0.08 ppm.

India has adopted standards or guidelines for several criteria pollutants, but has apparently not yet adopted one for ozone. Discussions are in progress, however, concerning development of an air quality index, which would necessarily include setting at least some sort of trigger level for ozone concentrations.

Some of the jurisdictions listed in Table 3 have also set standards or guidelines designed to be protective of forests and crops. The value is usually set as an AOT40 (average over time, relative to 40 ppb), which is defined as the sum of the differences between the hourly average concentration and 40 ppb for each hour for which the concentration exceeds 40 ppb. This sum is accumulated over the relevant growing season, and has units of concentration multiplied by time. The European Union has set a target value of $18,000 \mu\text{g/m}^3\cdot\text{hr}$ for 2010, and a longer term objective of $6,000 \mu\text{g/m}^3\cdot\text{hr}$ for 2020. These values refer to a three month growing season (May to July).

It may be noted, that in addition to the national or supra-national standards or guidelines described above, the World Health Organisation has proposed an ozone guideline of $120 \mu\text{g m}^{-3}$ (approximately 60 ppb) as an 8-hour average.

4. Control Measures

Measures introduced in Ontario (Ontario) to control the emissions of NO_x and VOC, which lead to production of ozone fall under the umbrella of the Clean Air Action Plan, which was introduced in 2004. The plan includes government regulation, while also encouraging non-regulated initiatives.

Specific initiatives under the Clean Air Action Plan include the following:

- Emission reductions by industry as laid out in the Industrial Emissions Reduction Plan, and embodied in O.Reg.194/05. Provisions are included for emissions trading
- Caps on NO_x emissions by Ontario Hydro thermal generating facilities, combined with the closure of the Lakeview generating station and planned phase-out of the remaining coal-fired generating stations in the province
- An inspection and maintenance program for motor vehicles (Drive Clean). Supplementing this program are the requirements for blending of ethanol into gasoline, plus fuel volatility and vapour recovery measures.

In summary, Ontario is developing, or has in place, requirements to reduce emissions of both NO_x and VOC, as is necessary for control of ozone. These measures are similar to those implemented in many other jurisdictions. It would be fair to say that the Law of Diminishing Returns is now operating in the province (as in the rest of the developed world). Alternatively stated, “the low-hanging fruit has been plucked”, and further advances will be increasingly more difficult and costly to achieve, requiring careful planning, and the full cooperation of the sectors responsible for the emission of NO_x and VOC.

However, it is worth noting that the transportation sector accounts for a significant fraction of the relevant emissions. Based on 2001 data, as reported in the Ontario Ministry of the Environment Transboundary Air Report (Yap et al., 2005), transportation accounts for 63% of NO_x and 29% of VOC (see Figure 2 for further details). To achieve further emission reductions overall, a high priority will have to be placed on this sector. The approach will necessarily have to be multi-pronged, incorporating at the least, continued measures to reduce the emissions of individual vehicles, with initiatives designed to reduce the number of vehicles on the road.

The latter component will involve active public participation, which will only work if it is

- Relatively convenient to travel by some means other than private light duty vehicles (e.g., improved public transit, urban planning which encourages other modes of transport)
- More costly and/or less convenient to travel by private automobile especially those of higher fuel consumption, e.g., parking surcharges in urban areas, surcharges on high fuel consumption vehicles, additional toll routes.

It is well known that London has imposed a congestion surcharge on vehicles entering the central area. Stockholm introduced such a surcharge for a trial period in 2006. During the trial traffic volumes entering the city were reduced by 20 to 25%, with concomittant reductions in queue times. Emissions in the city were reduced by about 14%, and a reduction by about 10% in traffic related injuries and fatalities was also noted (Algers et al., 2006). Following a referendum, it was decided to introduce the congestion charge as a permanent measure, starting in July 2007. Industrial and commercial response was initially strongly against the congestion charge, but is now assessed as neutral.

Rome has a similar, though less widely publicised charge in place for entry to the historic centre of the city. No private vehicles are allowed to enter the designated area, except those owned by residents of the area. These vehicles, as well as those of tradesmen who require vehicular access for their work pay a substantial licence fee.

5. Science Gaps

Research continues worldwide on improving our understanding of the production of ozone, and the way in which precursor emissions at one location are linked to concentrations at another. Accompanying improvements to the science is the continuing need to improve the data which support the analyses. As a particular example, there is an ongoing need to improve the quality and timeliness of emission inventory information.

It is not within the mandate of this review to carry out a detailed assessment of knowledge gaps which hold back or compromise assessment of problems related to ozone. Although now slightly dated, the NARSTO assessments (refs) provide an excellent, in-depth assessment of the state of science and knowledge related to ozone and to emission inventories.

However, some important areas in which improved knowledge is required have emerged in the course of the review, and may be summarised as follows:

- In discussing background ozone, it is important to be clear about what is meant by the term. In particular, distinction must be made between the concentration of ozone entering a region of concern as a result of emissions occurring outside the area, and the concentration at any point within the region of concern resulting from the combined effect of natural sources anywhere and anthropogenic sources outside the region .
- There is a clear need to refine estimates of background ozone, in particular resolving the discrepancies typified by the opposing views expressed in the work of Lefohn et al. (2001), Cooper et al. (2005) and Fiore et al. (2003). This includes the need for clarity on the role of the stratosphere as a source for troposphere ozone.
- The temporal patterns of background ozone need improved definition, on scales from diurnal upwards.
- An assessment is required of the relevance of temporal variability in background ozone in the calculation of health and other costs associated with ozone, specifically in relation to the separation of controllable and non-controllable impacts.
- Related to the assessment of costs of ozone damage, there is a continuing urgent need for improved data on health and other costs. Current estimates are in some areas based on very limited data.

6. Conclusions

It is now widely recognised that background ozone may form a significant part of the concentration experienced at many locations. This can be extremely important, since the background concentration is the level below which concentrations can not be reduced by local efforts alone. The background has both a natural and an anthropogenic component, and is a hemispheric phenomenon. Reported background ozone concentrations are similar for North America and Europe. The limited number of values reported for the southern hemisphere tend to be somewhat lower than for the northern hemisphere.

Although there is a considerable agreement between the concentrations reported by a number of different researchers, a significant discrepancy remains to be resolved. In particular, the role of the stratosphere as a source of ozone for the troposphere requires clarification. Resolution of the differences between the reports of high background ozone concentrations (e.g., Cooper et al., 2005), and those of much lower concentrations (Fiore et al., 2003) will require further research. The difference is important, because the background concentration is not controllable within a jurisdiction. Thus a low background concentration suggests that a large proportion of the total ozone concentration can be controlled, given the introduction of appropriate measures. For example, if the background is 25 ppb, local, anthropogenically produced ozone must be reduced to 40 ppb to achieve a standard of 65 ppb. However, if the background concentration is high, say 55 ppb, the local anthropogenic component will have to be reduced to 10 ppb, which is much more difficult.

Ozone lifted to levels above the boundary layer can be transported very long (intercontinental) distances (see Figure 3 for a schematic depiction of the vertical structure of the atmosphere). As a result, Asia, North America and Europe can all contribute to each other's background ozone concentrations. This means that, although agreement with ones immediate neighbours on emission reductions is very important in addressing the ozone problem, international agreements at much longer range must also be considered. In particular, the rapid industrial growth in India and China, and the accompanying increased precursor emissions, will need to be monitored and addressed.

7. Recommendations

- Public and media attention has swung significantly in the past year from photochemical smog (including ozone) to climate change as the major current environmental problem. It is important that the Ministry of the Environment not lose sight of the links between the two. Specifically, combustion of fossil fuels represents a major emission source for both phenomena. In fact, emissions from fossil fuel combustion also contribute to acid deposition, which is a subject which attracts little publicity, but remains important. In addressing air pollution it is therefore important to stress the cross-linking of benefits to be realised from abatement measures.
- Ozone continues to represent a hazard to human health in Ontario, and continued development of control strategies and measures is required. Such strategies will have to include continued tightening of emission limits for industrial sources, together with measures designed to decrease use of higher emitting sources and processes, e.g., provision of clean energy, improved public transport, disincentives to motor vehicle use, especially high emitters.
- When speaking of background ozone care must be taken that the term is defined appropriately. It is not possible to give a single definition because of different usages. In the Ontario context background ozone represents a boundary condition in mathematical modelling, but in other applications forms the uncontrollable portion of the atmospheric ozone in the province.
- It is recommended that the Ministry of the Environment continue to monitor, and support research on, the different views of background ozone as exemplified by Lefohn et al. (2001) and Cooper et al. (2005) on the one hand and Fiore et al. (2003) on the other. In particular, there is a need to understand why the within-domain background concentrations predicted by MOE modelling are higher than those predicted by Fiore et al. (2003).

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Appendix: Unit Conversions

Concentration expressed in parts per billion by volume, C_{ppb} is related to concentration expressed in micrograms per cubic metre, C , as follows:

$$C_{ppb} = (C \times 22.41 \times T \times 1013) / (1000 \times MW \times 273 \times p)$$

Where T is temperature (Kelvin)
 MW is molecular weight, and
 p is pressure (hectopascals)

For ozone at 20 C and one atmosphere pressure the conversion factor becomes

$$C_{ppb} = 0.50 \times C$$

Note that 1 part per million (ppm) = 100 parts per hundred million (pphm)
 = 1000 parts per billion (ppb)

Table 1. Background Ozone Concentrations at Several Locations

Location	Concentration (ppb)	Reference	Comments
Mainly North America: Low Elevation			
US	Mainly 25 to 35; some higher	Fiore et al. (2002)	Determined from modelling using GEOS-CHEM
US	Mainly 15 to 35; some to 45	Fiore et al. (2003)	Determined from modelling using GEOS-CHEM
US	35 ± 10	Altshuller and Lefohn (1996)	
US, Europe	50 – 60	Lefohn et al. (2001)	Stratospheric input
US, Canada, plus some European	28 to 50	Lefohn et al. (2001)	Measurements at “clean” sites
California	32 to 44	Jaffe et al. (2003)	Six coastal sites
Europe: High Elevation			
Switzerland	33 to 50	Bronnimann et al. (2000)	Concentration increased with altitude. Highest on Jungfrauoch (3500 m)
Mt. Cimone, Italy	55 ± 7	Bonasoni et al. (2000)	High elevation (2165 m)
Germany, Switzerland	30 to 60	Naja et al. (2003)	High altitude locations. Back trajectory analysis
Europe: Low Elevation			
Mace Head, Ireland	39.3 ± 6.0	Simmonds et al. (2004)	Coastal site
Europe	30	Auvray & Bey (2005)	Determined from modelling using GEOS-CHEM
Europe	30 to 35	ACCENT 2006	Review
Gozo, Malta	52.2	Nolle et al. (2001)	Background for Malta, but not for Europe
Scandinavia	25 to 40	Lindskog and Kindbom (2001)	Measurements made at remote sites in N. Scandinavia
Europe	30 to 37	Scheel et al. (1997)	From analysis of measurements at several locations
Eastern and Oceanic: High Elevation			
Hawaii	“Substantial” (e.g., 60)	Cooper et al. (2005)	Stratospheric input
Happo, Japan	44.4 ± 3.6	Pochanart et al. (2004)	Mountain site (1840 m)

Multiple Locations and Reviews			
Several sites globally	25 to 50	Oltmans et al. (2006)	Measurements at remote sites
Review	40 to 80	Lefohn (2001)	
Review	14 to 47	Vingarzan (2004)	Review of published values
Review, US locations	25 to 45	Fiore et al. (2003)	

Table 2: Reported trends in background ozone

Rising trend (per year)	Time Period	Location	Reference and Comments
0.5 to 2%	Various	Various	Vingarzan (2004)
Little significant change	1988 – 1996	Northern Scandinavia	Lindskog & Kindboom (2001)
0.5 ± 0.36 ppb	1986 – 2004	California	Jaffe et al. (2003)
0.49 ± 0.19 ppb	1987 – 2003	Mace Head, Ireland	Simmonds et al. (2004)
5 to 10 ppb over 30 years	1970 – 1999	Switzerland, Germany	Naja et al. (2003)
1 ppb	1997 – 2000	Gozo, Malta	Nolle et al. (2001)
0.8 to 1.3 ppb over 9 to 11 years	1988 – 1998	Jungfraujoch, Switzerland	Bronnimann et al. (2000)
0.18 ± 0.04 ppb	1990 – 2001	Mace Head, Ireland	Carslaw (2005)
Up to 0.5 ppb	1973 – 2004	Remote locations	ACCENT (2006)

Table 3. Ozone Standards/Guidelines for Selected Jurisdictions

Jurisdiction	Value	Averaging Period (hr)	Description
Canada	65 ppb	8	Fourth highest annual value averaged over three years. To be achieved by 2010
United States	0.08 ppm	8	Fourth highest annual value averaged over three years.
Mexico	0.110 ppm	1	Daily maximum
European Union	$120 \mu\text{g m}^{-3}$ (60 ppb)	8	Not to be exceeded more than 25 times per calendar year, averaged over three years. To be achieved by 2010
Japan	0.06 ppm	1	
China	0.12 mg m^{-3}	1	This limit is for Class I (residential) areas. Higher limits are set for Class II and Class III (industrial) areas
Australia	0.08 ppm	4	
India	India have not yet set a standard or guideline for ozone		

Table 4. Projected background ozone concentrations for future years

Year	Low end of range (ppb)	Average (ppb)	High end of range (ppb)
2040	34	43	50
2060	36	54	70
2080	39	63	85
2100	40	62	84

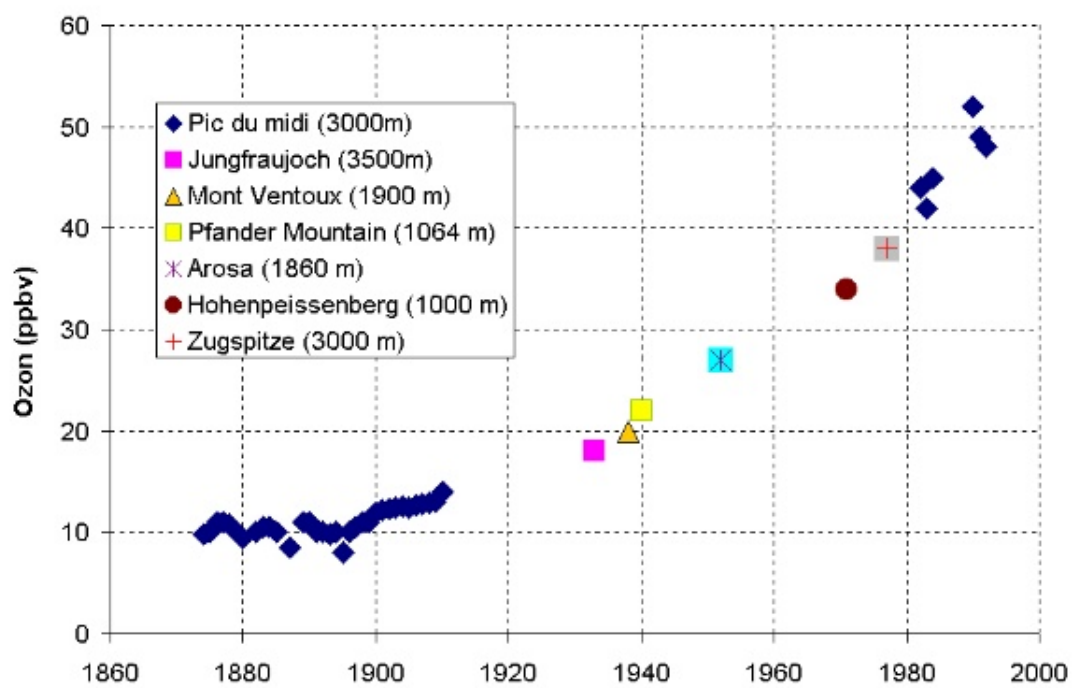


Figure 1. Tropospheric ozone trends (Gros, 2006, adapted from Marenco et al., 1992)

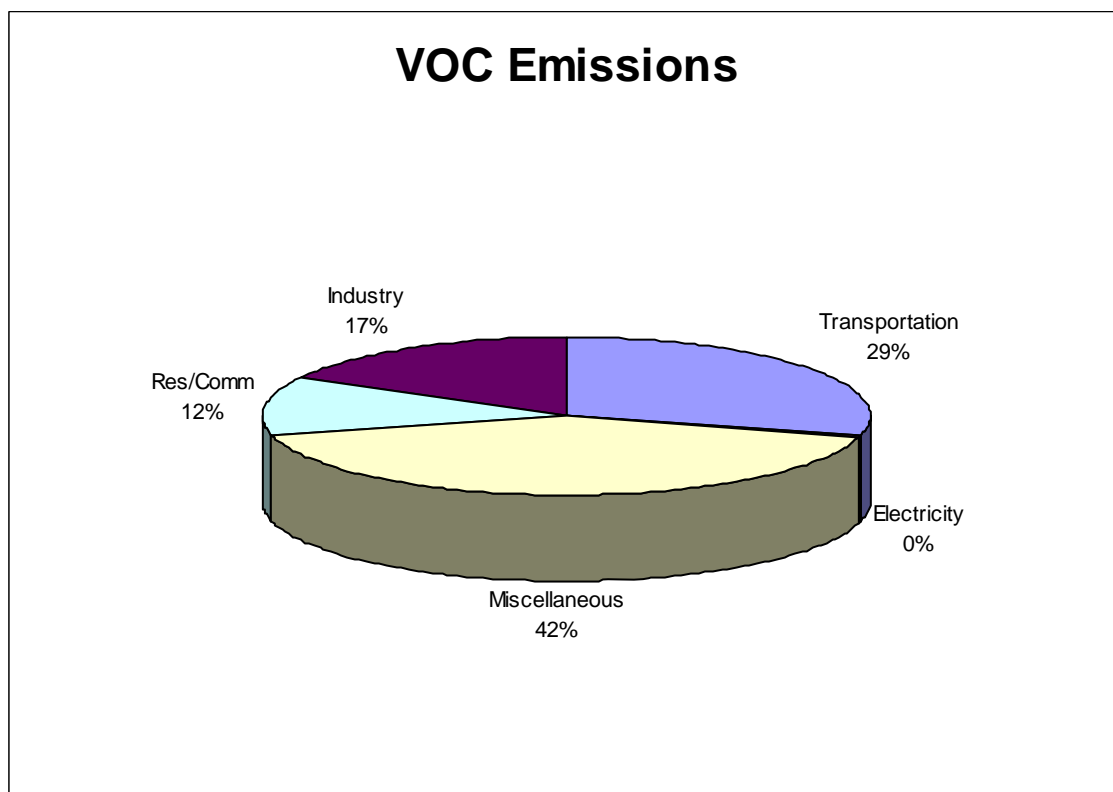
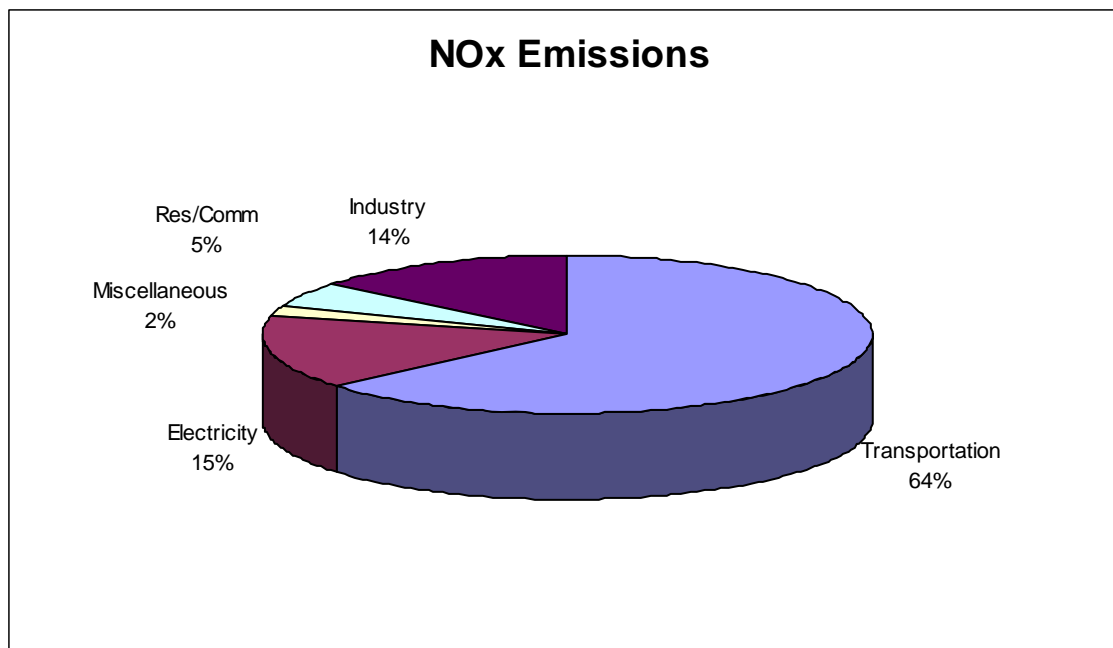


Figure 2. Ontario NO_x and VOC emissions by sector (from Yap et al., 2005)

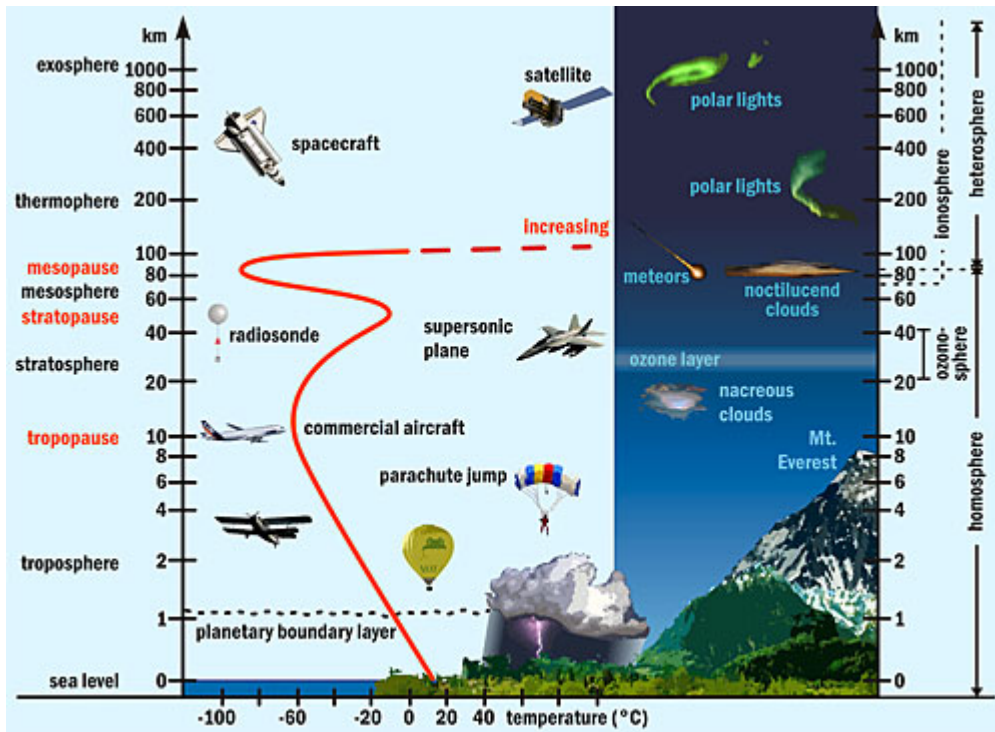


Figure 3. Vertical Structure of the Atmosphere (from Kowama GPS, <http://www.kowoma.de/en/gps/additional/atmosphere.htm>)